Two Dimensional ^{89}Y NMR Study of Vortex Dynamics in $YBa_2Cu_3O_{7-\delta}$

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(Received 6 April 1995)

We have performed two dimensional ^{89}Y NMR experiments on a powder sample of $YBa_2Cu_3O_{7-\delta}$ around the vortex lattice melting temperature in a magnetic field. Local motion of vortices with small amplitude is observed; however, long range diffusive motion of the vortices appears to be absent over the time scale of 10 s.

PACS numbers: 74.25.Nf, 74.40.+k, 76.60.Jx

There has been much interest recently in the vortex liquid to solid (or vortex glass) transition and the effects of vortex pinining in high temperature superconductors [1]. An essential question regarding vortex dynamics, which remains unclear, is the magnitude of the time and length scales for the vortex fluctuations to occur. These basic parameters of vortex dynamics are related to the dissipation processes and the quasiparticle excitations in vortex cores. Although extensive investigations using the transport measurements [2] have provided deep insight into the thermodynamics of the vortex phase transition, they do not probe the detailed vortex motion directly.

We address this problem with nuclear magnetic resonance (NMR) experiments. The essence of this general approach is that NMR probes the magnetic field distribution and fluctuations that exist in the mixed state of a type II superconductor, such as $YBa_2Cu_3O_{7-\delta}$. The formation of a vortex lattice produces a peculiar magnetic field distribution, and this was first recognized 40 years ago by Abrikosov [3]. Subsequently, such field distribution was observed in NMR experiments [4] for elemental, low T_c superconductors.

This Letter describes an NMR experiment that measures the correlation function of magnetic field distribution as the vortices move. This experiment, in general, will unambiguously determine the amplitude and the time scale of vortex fluctuations. The principle of this experiment is as follows. Suppose one follows an individual nuclear spin. The resonance frequency of this spin is proportional to its local magnetic field at the time of detection. The experiment involves measuring the resonance frequency at the beginning of the experiment, ω_1, and at the end of the experiment, ω_2, separated by an evolution period, τ_e. If the local magnetic field does not change during the evolution period, for example, $\tau_e < \tau_c$ (the correlation time of vortex dynamics), one would observe $\omega_1 = \omega_2$. If the magnetic field does change during τ_e, then $\omega_1 \neq \omega_2$. In an actual experiment, one obtains the ensemble average of such correlations. In a plot of the correlation function with ω_1 and ω_2 as variables, the off-diagonal signals indicate a change of the local field, and thus vortex motion. Such an experiment is considered two dimensional (2D) since two independent frequencies are measured directly [5].

In this Letter, we report two dimensional ^{89}Y NMR experiments on $YBa_2Cu_3O_{7-\delta}$. We found that the long range diffusive motion of vortices is absent above and below the vortex melting temperature, within a time scale of 10 s. We will show that this result is independent of theoretical models for vortex dynamics.

A powder sample of $YBa_2Cu_3O_{7-\delta}$ was mixed with epoxy resin (Stycast 1266, Emerson and Cumming, Inc.) at a filling fraction of 20% by volume. The composite was cured in a 4.2 T magnet to produce stable c-axis alignment of the crystallites. The angular dependence of the ^{89}Y NMR spectra shows that the sample is essentially fully aligned. The superconducting transition of the sample is measured to be 87 K at 9.4 T field.

The NMR experiments were performed at a field of 9.4 T magnet using a Bruker AM-400 spectrometer. Temperature was maintained stable within 0.1 K using a flowing helium cryostat (CF1260N, Oxford Instrument, Inc.). We have examined the ^{89}Y NMR spectrum around T_c, and the linewidth was found to increase below T_c. This behavior has been observed previously by Markert *et al.* [6] and Brom and Alloul [7], and the present result is consistent with theirs over the temperature range explored. The broadening is a measure of the magnetic field distribution due to the formation of the vortex lattice. Another interesting phenomenon that Markert *et al.* [6] discovered is that the spin-spin relaxation rate becomes enhanced right below T_c. This was later explained by Suh, Torgeson, and Borsa [8] to be an effect of vortex dynamics. The present measurements also show a similar behavior.

A standard three pulse sequence [9] was used for the 2D experiment. The generic pulse sequence is noted below:

$$\pi/2_{\phi_1} - t_1 - \pi/2_{\phi_2} - \tau_e - \pi/2_{\phi_3} - t_2(\text{detection}). \quad (1)$$

Here, $\pi/2$ indicates a radio frequency (rf) pulse that rotates the ^{89}Y spin by 90°. The subscripts ϕ represent the phases of each rf pulse, and proper sequence of these rf phases removes artifact signals including various forms of coherent noises. The typical $\pi/2$ pulse is 15 μs.

Let us analyze the above pulse sequence in more detail. Assume ϕ_1 and ϕ_2 are set to be equal. During t_1, the nuclear spins precess under the local field at each nuclear site. The relative phase accumulated is $\omega_1 t_1$. Since ω_1

 0031-9007/95/75(10)/2008(3)$06.00

may be different for each nuclear spin, their corresponding phase is stored along the static field (**z**) by the second rf pulse. Right after the second rf pulse, the magnetization m along **z** is

$$m_z(t_1) = \int d\omega_1 \, P(\omega_1) \cos(\omega_1 t_1) \,, \qquad (2)$$

integrated over the spectrum, $P(\omega_1)$. During the evolution time τ_e vortices may diffuse and the local field may change. At the end of τ_e, a third rf pulse is applied to detect the nuclear spin procession frequency, at a new local field ω_2. A typical experiment takes 128 equally spaced t_1 values and 128 data points in the detection, t_2. The detected signal can expressed as

$$m(t_1, t_2) = \int d\omega_1 \, d\omega_2 \, \chi(\omega_1, \omega_2) \cos(\omega_1 t_1) \exp(i\omega_2 t_2) \,. \qquad (3)$$

$\chi(\omega_1, \omega_2)$ is the probability for a spin at frequency ω_1 and ω_2 at the beginning and the end of τ_e. The data are then a matrix of the dimension 128 by 128, and a two dimensional Fourier transform is applied to the matrix to obtain the frequency correlation function, $\chi(\omega_1, \omega_2)$. The nuclei that do not change their resonance frequency at the beginning and the end of τ_e appear on the diagonal line in $\chi(\omega_1, \omega_2)$. Those nuclei that experience a change in the resonance frequencies will produce an off-diagonal signal. Thus the appearance of such an off-diagonal signal is the signature of vortex diffusion. For unresolved resonances, a 2D contour map becomes circular in the limit of long range diffusion.

In Fig. 1, we show our results from the 2D experiments. The contour plots show the frequency correlation function at 88 K (a) and 82 K (b). The vortex melting temperature is estimated to be 77 K for $YBa_2Cu_3O_{7-\delta}$ at the magnetic field of 9.4 T, extrapolated from the data in Ref. [2]. In the normal state, the ^{89}Y linewidth is due primarily to material inhomogeneity and we do not expect to observe exchange or diffusion to occur at 88 K, and this is confirmed by noting the data in Fig. 1(a) which shows only diagonal features and no off-diagonal signal.

The ^{89}Y linewidth in the superconducting state was found to increase considerably compared to that of the normal state, due to the formation of the vortex lattice. This can be seen from the spectrum of the spin echo and also from the 2D experiment shown in Fig. 1(b) measured at 82 K. We have performed 2D experiments for τ_e = 0.1, 1, and 10 s at this temperature. No significant off-diagonal signal is observed in any of these experiments. Comparison of the experiments with different τ_e shows no decrease in the integrated signal intensity for longer τ_e, and this indicates quantitatively that the lack of off-diagonal signal is not due to poor signal-to-noise ratio in those areas.

We have also performed the 2D experiments down to 76 K and obtained essentially the same result. Experiments at lower temperatures are too time consuming because the long ^{89}Y spin-lattice relaxation times preclude extensive signal averaging.

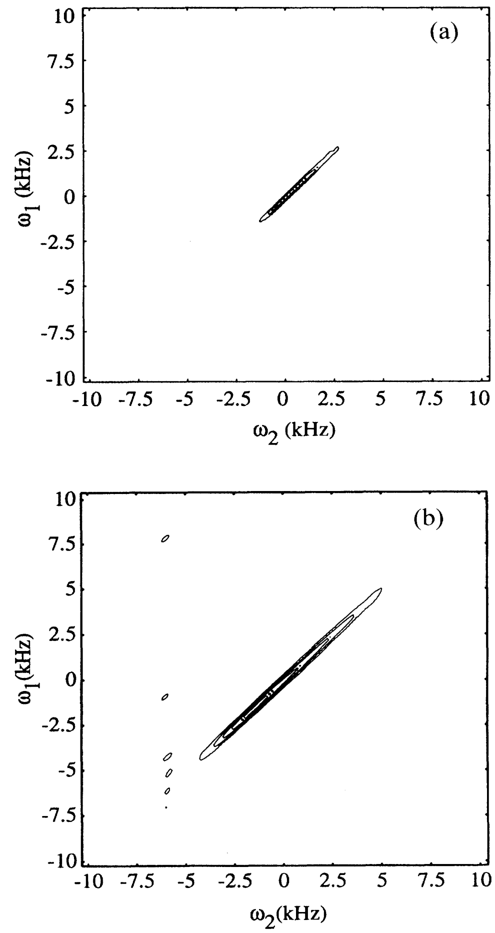

FIG. 1. Two dimensional contour plots of the field correlation functions obtained by Fourier transform in both t_1 and t_2 dimensions. Both axes show the frequency offset from the spectrometer frequency. Measurements were performed at (a) 88 K and (b) 82 K, and the magnetic field is 9.4 T. For both measurements, τ_e = 1 s and no line broadening was applied. Contours are at 90%, 70%, 50%, 30%, and 10% of the maximum signals. The circles around $\omega_2 = -6$ kHz are 10% contours due to noises.

The ^{89}Y linewidth originates from the spatial distribution of the magnetic field in the vortex lattice. The field is the highest at the vortex cores, so that the high frequency part of the ^{89}Y spectrum corresponds to the nuclei near vortex cores. The saddle points, midway between every two adjacent vortices, produce a peak in the field distribution and thus the ^{89}Y spectrum. For vortex motion with displacement on the order of intervortex spacing, $\approx$160 Å for our field, one should observe signals significantly away from the diagonal ridge. Thus, *the absence of such off-diagonal signals indicates the absence of large amplitude vortex motion and long range vortex diffusion over a time period of 10 s.*

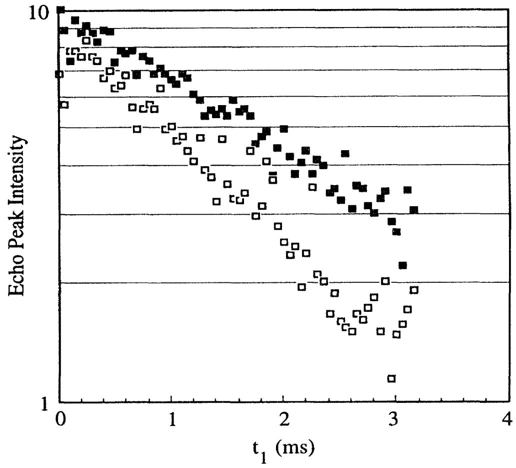

FIG. 2. The peak intensity of the stimulated echo as a function of t_1 for evolution time of 0.1 and 10 s, measured at 82 K.

The effect of small amplitude vortex fluctuations, however, is manifested more clearly in Fig. 2, where we show the peak intensity of the signal as a function of t_1 for $\tau_e = 100$ ms and 10 s. The faster signal decay of $\tau_e = 10$ s indicates a slight broadening of the 2D spectrum along ω_1. This small amount of extra broadening indicates vortex motion of small amplitude during the evolution period of 10 s. This increase in $\Delta\omega_1$ is similar in effect as compared to that observed from the spin-spin relaxation rate [8].

However, the present results seem to be at odds with the extensive transport measurements that show an abrupt resistive transition in the magnetic field for untwinned single crystals [2], even though the short range vortex fluctuations may also give rise to dissipation determined in transport measurements. It has been shown [2] that vortex pinnings may change the resistive transition considerably compared to measurements on the pure, untwinned single crystals. Typically, the transition becomes much broader and smooth for samples with high pinning density. It is very likely that our powder sample has sufficiently high pinning density so that the melting transition and the vortex motion might have a different character from that observed on untwinned single crystals.

Various time scales play important roles in interpreting different NMR experiments. For example, the spectral linewidth, $\Delta\omega$, may be affected by motion on the time scale of $1/\Delta\omega$. The spin-spin relaxation rate, $1/T_2$, will be affected if motion is fast during the time interval, T_2. Both effects can be categorized as motional narrowing. The spin-spin relaxation experiments of Suh, Torgeson, and Borsa [8] are sensitive to short range fluctuations of vortices (over a small fraction of intervortex spacing) within a time scale of a few milliseconds. In their analysis, a linear field gradient and random diffusion processes were assumed. These assumptions are likely to be incorrect for larger amplitude fluctuations. For the 2D experiment described above, one can monitor motion during τ_e, and τ_e can be varied independently [10]. The most important advantage of the 2D experiment is that one obtains the amplitude of the motion directly, without any theoretical modeling. Essentially, this experiment can tell us unambiguously whether vortices move or not during τ_e, on the distance scale of the intervortex spacing.

In summary, we describe a new method to study the vortex dynamics through the correlation function of magnetic field distribution. The experiments have shown unambiguously that long range diffusion of vortices does not happen in a time scale up to 10 s, above or below the melting temperature. The vortex fluctuation amplitudes are limited to a small fraction of the intervortex spacing. However, it is unclear if this is due to the effects of pinning present in the powder sample or it is intrinsic to the vortex lattice in $YBa_2Cu_3O_{7-\delta}$.

We thank D. A. Wright, J. P. Emerson, and N. E. Phillips for providing the $YBa_2Cu_3O_{7-\delta}$ sample, and Dr. J. Shore, J. A. Reimer, A. Pines, and W. P. Halperin for discussions. Y. Q. S. acknowledges the Miller Research Fellowship from Miller Institute for Basic Research in Science. This work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences Division of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

[1] D. J. Bishop, P. L. Gammel, D. A. Huse, and C. A. Murry, Science **255**, 165 (1992); some theoretical issues are reviewed in G. Blatter *et al.,* Rev. Mod. Phys. **66**, 1125 (1994).

[2] H. Safer *et al.,* Phys. Rev. Lett. **69**, 824 (1992); W. K. Kwok *et al.,* Phys. Rev. Lett. **69**, 3370 (1992).

[3] A. A. Abrikosov, Zh. Eksp. Teor. Fiz. **32**, 1442 (1957) [Sov. Phys. JETP **5**, 1174 (1957)].

[4] P. Pincus, A. C. Gossard, V. Jaccarino, and J. H. Wernick, Phys. Lett. **13**, 21 (1964); W. Fite and A. G. Redfield, Phys. Rev. Lett. **17**, 381 (1966); A. G. Redfield, Phys. Rev. **162**, 367 (1967).

[5] This and other types of multidimensional NMR techniques have been used extensively in a wide area including the study of molecular motion, chemical exchange, and diffusion, and an excellant overview of the field can be found in R. R. Ernst, G. Bodenhausen, and A. Wokaun, in *Principles of Nuclear Magnetic Resonance in One and Two Dimensions* (Clarendon Press, Oxford, 1994).

[6] J. T. Markert, T. W. Noh, S. E. Russek, and R. M. Cotts, Solid State Commun. **63**, 847 (1987).

[7] H. B. Brom and H. Alloul, Physica (Amsterdam) **177C**, 297 (1991).

[8] B. J. Suh, D. R. Torgeson, and F. Borsa, Phys. Rev. Lett. **71**, 3011 (1993).

[9] G. Bodenhausen, H. Kogler, and R. R. Ernst, J. Magn. Reson. **58**, 370 (1984).

[10] This evolution time τ_e should be shorter than the ^{89}Y spin-lattice relaxation time.